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From: Bernard Frank

Subject: RI-CLIMATIC RELATIONS-Precipitation-Use of Tritium

Notes on Talk by Dr. W. F. Libby (AEC) on
Natural and Artificial Tritium and its Use in Hydrology

Dr. Libby spoke on the above subject at the Cosmos Club on October 20, 1955, under the auspices of the Washington Academy of Sciences. His main theme was that cosmic radiation produces a measurable quantity of radioactive tritium (T) from hydrogen atoms in the upper 1/10 of the atmosphere (above 80,000 feet), and that because of the relatively long half-life of T (12.5 years) part of the quantity produced filters down into the lower atmosphere, coming to earth via precipitation, thus making it possible to determine sources of precipitation, and the age of surface and underground waters.

In order to obtain a more accurate idea of the nature and implications of Dr. Libby's remarks, two articles[1/] published prior to his talk were also referred to subsequently, so that what follows represents an attempt to synthesize the contents of the talk and the articles.

T--an isotope of hydrogen--is dispersed through the atmosphere and waters of the earth. Its total amount is about two pounds. Of this amount, all the inland waters and the atmosphere contain about one ounce, of which these waters have about one-third. "However, its radioactivity and modern analytical techniques make it possible to measure accurately the T in a single gallon of river water. It makes a fine measure for determining the age of plants and exploring the weather."

The radioactivity (beta ray emanations) accompanying decay of the T atoms can be measured in air or in water, whether as rain, snow, surface or underground waters. Thus, calculations can be made of the age of this moisture in the air mass or in bodies of water, on the basis of the amount of radioactive decay, as compared with the original rate.

1/ Libby, Willard F. Tritium in nature. Sci. American 190(4): 38-42. 1954.

Kaufman, Sheldon and W. F. Libby. The natural distribution of tritium. Physical Review 93(6): 1337-1344. 1954.

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Most of the T made by cosmic rays must form water in the atmosphere. This water probably falls and reaches the oceans before any appreciable fraction of the T is lost by radioactive decay; its half-life indicates that the rate at which the oceans receive it must be nearly equal to the total rate of production. In calculating the amount that falls into the oceans directly as rain and the amount that flows in from the Continents by way of rivers, we must take into account the varying intensity of cosmic radiation with latitude; about 4 times as great at the latitude of Chicago, for example, as at the equator. Since T probably mixes vertically faster than it does horizontally, we should expect the T content of rains to show corresponding variations with latitude. Calculations indicate that the average T content of sea precipitation in the vicinity of Bergen, Norway, is 0.54. Multiplying by the total annual ocean rainfall--a little over 30 inches--the total T falling directly into the ocean is thus obtained.

To add the T flowing in from the rivers, Mississippi River measurements were taken and corrected for the latitudinal factor giving a world-wide average of 3.3 for the T content of land rainfall. The average total runoff from all continents into the oceans is about 11.4 inches per year. These figures permit calculating the amount of T entering the sea from the rivers. From the above can be concluded the average rate of production of T over the whole earth, namely, about 0.1 T atoms per square centimeter per second. The production at any location can also be computed.

An evaluation of precipitation and lake and river waters in the Mississippi valley discloses that the T assay of ocean rains is much lower than that of inland precipitation because: (1) Water which rains back into the sea has been in the air for a much shorter time and therefore has had a shorter exposure to cosmic rays than that which moves in over the land before it precipitates. (2) Water evaporated from the sea should have no appreciable amount of T because the isotope is so diluted in the ocean. (3) On the other hand, rain falling on the land is not diluted and hence should accumulate T in each successive reevaporation and fall. Thus Mississippi valley rains have 6 or 7 times as high a T content as rains on Hawaii or the Norway coast. In general, the closer to the sea coast, the less T in the rain. More observations lead us to expect that waters on the western slopes of continents will have less T than those on the eastern slopes because of the prevailing westerly winds. The wide variability of Chicago rains (from 1 to 66 T atoms on the index) appears to indicate that the water in some of the rains had been rained out on the land and reevaporated many times. The Chicago T fall also seems to have a seasonal cycle--generally highest in the fall and winter months. This seems to correlate with the fact that in those months Chicago gets most of its moisture from the distant Pacific and in spring and summer from the Gulf of Mexico. Knowing the rate of production, we can write an equation making it possible to calculate how long moisture has been in the air--from the time it is evaporated from the ocean until it falls as rain. Based on a rough estimate of the average amount of moisture in the atmosphere and a T assay of oceanographic rain, calculations indicate that water stays in the air over the ocean an average of about 9 days.

The richest natural sources are rain and snow. (By contrast, the earth's crust represents a very poor source, as a layer of igneous rock one km. thick has a T production rate of only 0.001-T atoms per cm^2/sec. as against a likely cosmic ray production rate of 0.1 or higher. Similarly, production in sea water is very negligible.)

The possibilities of utilizing T for meteorological purposes derives from the fact that the test for T in a given storm depends upon correlating its concentration and stage of decay with the trajectory of the air mass and its water content.

"Once the T production rate (Q) has been evaluated, the expected value of T, that is, the tritium concentration in water in an air mass, can be calculated in terms of the amount of water w (meters per cm^2) and the time r, in years, elapsed since the water evaporated from the oceans. For a given latitude we would write

$$Tw = 4.7Qr$$

"Example: Water content of average air mass traveling over the Hardangervidde Plateau in Norway is 3 g per cm^2, r = 0.024 years, or about 9 days average.

"Table 4
Average T content for precipitation in the northern hemisphere

Location	Latitude (geomagnetic)	Precipitation (meters/ year)	T content (T atoms/ 10^{18} H's)
Lake Michigan	~54°N	0.80	7.7 ± 0.3
Mississippi Basin	48°N	0.76 (0.19 runoff)	5.2 ± 0.2
Naples, New York (wine)	~54°N	1.0	5.8 ± 0.3
Rhone Valley, Tain, Drome, France (wine)	~54°N	0.8	3.4 ± 0.3
Bordeaux, France (wine)	~54°N	0.8	4.3 ± 0.3
Lake Mösvann, Norway	55°N	0.9	0.8 ± 0.1

"The much larger values of the T concentration found in the rain and runoff over the land masses (Table 4) as contrasted to sea rains and snows undoubtedly are due to longer exposure of these waters to atmospheric T. By formula it is determined that the water flowing into the Gulf of Mexico and New Orleans left the ocean about 11 months earlier. More accurate values of w should be used to improve this calculation.

"A rainfall value for Lake Michigan of 7.7 is calculated by use of a factor of 4.5 for the storage and hydrologic effects. An equation is used to derive the expected ratio of the tritium content of rain to that of lake water as 4.5."

As the above passages indicate, studies of rainfall show wild fluctuations in storm amounts and in the concentration of T in such rains (no two storms being alike). The concentration of T is an inverse function of the amount of water in the air. The average amount of water in the air has a sinusoidal variation ($y = a \sin x$) throughout the year. Since the prevailing winds over the northern hemisphere are west to east, the air mass concentration--and the corresponding concentration of tritium--rises from a factor of 1 over the Pacific Ocean and immediately adjacent California coast, to a factor of 9 over the Atlantic coast. The concentration again drops in air masses lying over the Atlantic Ocean itself. (See illustrative chart on p. 41 of Sci. American article cited.)

Ocean rains are very different from land rains in respect to T content. As previously stated, rainfall over the land has little chance of dilution by ocean water because it results largely from moisture evaporated from the land surface, so that the tritium becomes more concentrated, and thus the ratio increases. Little change in tritium concentration occurs during a given rain. (Incidentally, Dr. Libby said that during the coming International Geophysical Year it is planned to obtain a large series of samples throughout the world with the help of meteorologists.)

Ground-Water Movement

The source and age of ground water are determined by measuring the T concentration either in existing wells or specially drilled wells. (Latitude has to be considered since the concentration also varies with this factor.) For example, tests of 300-foot deep wells in Urbana, Ill., showed that Urbana is using fossil water. This supply is not being replenished at any finite rate and therefore the supply is definitely exhaustible. Studies of selected wells over a large area in Nebraska where the water was under pressure under an impervious clay cap also showed very little tritium in evidence. This means that the water is very old and not easily replenishable, so here again this supply appears doomed. (It costs about $150 to take a sample for such determinations. This is a commercial rate, and commercial concerns are doing this work.)

The storage time of ground water is also determinable by measuring the T content. Of course, trained hydrologists are needed to interpret the results. A number of European rivers were measured in September 1953. The relative values of tritium concentration ran from 1+ to 2+. (2.0 equals the California level.) The Mississippi River is 6 (1.0 being the concentration over the Pacific Ocean). At St. Louis in January '53, the Mississippi River had a concentration of 5.6.

Hot Springs

The natural hot springs nearly all have T except those that are very high in salts. For example, tests at Wilson Spring, Arkansas, Lassen Park Geyser, and Minerva Spring in Yellowstone National Park indicate that the water is of recent age. In other words, rain water is continually seeping down into the hot subsurface strata.

Ocean water has a much lower T content than ocean rain--one-fifth as much (see preceding text). There is little mixing of water in the sea. A discontinuous layer (the thermocline) occurs at about 100 meters. No T is found below it. The implication for atomic waste disposal is that such disposal should be satisfactory in deep ocean areas below 100 meters. All plant life in the ocean occurs within this thin skin of 100 meters. The deep ocean life below this depth lives off the material which drops down in.

Changes in T concentration permit determination of the rate at which water moves to the sea in surface and ground-water streams.

Radioactivity went up 10 times on the Atlantic and Pacific Oceans following the H-bomb test in the spring of 1954. Now we can date and locate the origins of the world's air masses. Incidentally, little T has been found in the air below the equator, i.e., in the southern hemisphere.

Tests show Crater Lake turns over every 6 months. Studies also show that the average ground-water depth in the northern Mississippi drainage basin is 0.5 meter.

Applications

Tritium is cheap and is easy to "mark" large amounts of water with it; 10^{10} tons of water can be marked for $20,000 worth of T. The biggest cost is in making the heavy water. The water samples are all distilled by electrolytic processes before being tested.

The whole atmosphere of the world has only 10 grams of T. Most of the remaining 2 pounds is in the ocean "and so diluted as to be beyond detection." So the waste of a very little amount is enough to upset the results of measurement.

Discussion

Concerning precipitation over the North American Continent, comments by meteorologists at the meeting brought out that Libby's findings show a discrepancy with the now accepted theory that precipitation over the land is derived from ocean water, rather than from land evaporation. Dr. Libby expressed disagreement with these findings, as his studies and those of his associates show that, on the basis of tritium concentrations, precipitation over the land is derived from water which has previously been precipitated on the land and reevaporated into the atmosphere.

Don Pack of the Scientific Services Division of the Weather Bureau, who also attended the lecture, informs me that Dr. Libby's statements were too sweeping. The sampling technique is very new, he said, and leaves much to be desired by way of accurate measurements. The findings presented by Libby are based on only a few measurements. Perhaps if more data were obtained, the results would be quite different.

Pack also said, in commenting on the low T content of ocean water reported by Libby, that Libby didn't mention the slow overturning of sea water, whereby the lower layers rise up, bringing low T content water to the

surface. Such a process, he said, occurs all along the Pacific Coast of the U. S. Libby did not make clear where the samples came from--the surface, or deeper layers.

He pointed out that the factor and effects of atmospheric air circulation didn't receive adequate consideration. Meteoroligists did not participate in the tests or analyses.

On the other hand, Pack felt that the statements on ground water seem to be based on firmer evidence than that applying to the meteorological aspects.

Note: As to Libby's statement that most of the interior land precipitation originates from water evaporated from the land, I would like to call attention to a statement by I. R. Tannehill on p. 87 in Water, Yearbook of Agriculture 1955:

> As world populations increase, some parts of the problem assume tremendous importance. In the United States the rainfall climates range from the superhumid in some Eastern and Southern States to the dry climates of large regions in the West and Southwest. We deal with the problems in various regions by irrigation, drainage, flood control, soil conservation, and similar practices. In a vast region, however, where rainfall normally is adequate for agriculture (though barely so in some marginal areas) it is so variable that frequently extremes cause minor or major disasters. In some degree, the situations are made more dangerous by man's actions, such as cultivation and denudation, which eliminate a part of Nature's stabilizing controls and contribute to disaster, at least in local areas.

Perhaps there is some connection between the two statements. It is hoped that the additional measurements and analyses by meteorologists planned for this year will develop further substantial evidence on this quite important issue.

Bernard Frank